

Universal Heat Transport in Sr_2RuO_4

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We present the temperature dependence of the thermal conductivity $\kappa(T)$ of the unconventional superconductor Sr_2RuO_4 down to low temperatures (~ 100 mK). In $T \rightarrow 0$ K limit we found a finite residual term in κ/T , providing clear evidence for the superconducting state with an unconventional pairing. The residual term remains unchanged for samples with different T_c , demonstrating the universal character of heat transport in this spin-triplet superconductor. The low-temperature behavior of κ suggests the strong impurity scattering with a phase shift close to $\pi/2$. A criterion for the observation of universality is experimentally deduced.

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Through the study of the cuprate superconductor in the last decade, a notable progress has been made in an understanding of the thermodynamic and transport properties of quasi-particles (QP) in unconventional superconductors. One of the important findings is the novel phenomenon called the universal transport, pointed out first by Lee [1]. In an unconventional superconductor with nodes in the superconducting (SC) gap, non-magnetic impurities suppress the transition temperature (T_c) and induce the finite density of QP states at the Fermi level with the energy width γ_{imp} [2]. The presence of impurities increases both the QP density and the impurity scattering rate, which completely cancel each other [3,4]. In this case the QP transport, such as thermal conductivity (κ), restores the temperature (T) dependence typical for the normal state (e.g. $\kappa \propto T$) [5] and becomes independent of impurity concentration. The magnitude of the residual term in κ/T depends only on the QP spectrum near the gap nodes, providing a useful tool for studying the SC order parameter [4].

Experimentally, the universal residual term in κ/T at $T \rightarrow 0$ K has been reported in optimally-doped high- T_c cuprates $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$ [6] and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ [7,8]. Their residual values are consistent with the d -wave SC gap structure [9]. For non- d -wave gap symmetries, the universal conductivity has never been observed. In contrast, the extrapolated κ/T at $T \rightarrow 0$ K of the spin-triplet superconductor UPt_3 [10,11] rapidly increases with the density of defects, showing no universal behavior, and the residual term seems to vanish in zero-disorder limit [11]. The origin of the lack of universality is still unclear, similar to the case of under-doped cuprates [12], and it is strongly desirable to extend studies to other unconventional superconductors.

In this respect, of special interest is the study of Sr_2RuO_4 , a unique quasi-two-dimensional (Q2D) spin triplet superconductor [13,14]. This material has a layered perovskite structure [15] and its electronic state is well described in the Landau-Fermi liquid model [16]. The SC state of Sr_2RuO_4 ($T_c = 1.5$ K) is chiral with a

broken time reversal symmetry [17] and its T_c is rapidly suppressed by non-magnetic impurities [18,19]. Views on the SC gap symmetry of Sr_2RuO_4 changed in time. Early theoretical prediction of the fully gapped p -wave chiral state [20] did not find support in recent experiments, suggesting the SC gap with line nodes [21–24], likely running parallel to the conducting plane [25–27]. Several new theoretical models have been proposed [28–31] to explain the experimentally observed "triplet-chiral-nodal" SC order parameter.

The thermal conductivity measurement of Sr_2RuO_4 down to 20 mK was reported by Suderow *et al.* [32]. This study was restricted to strongly disordered samples with $T_c < 1$ K. Measurements with high quality samples, with $T_c > 1.4$ K, have been performed only down to 0.3 K [24–26], which is insufficient for the estimation of the residual term.

In this Letter we report the systematic study of the in-plane thermal conductivity of Sr_2RuO_4 down to 100 mK on samples with widely varying quality, with T_c from 0.7 K to 1.5 K. We found a universal behavior of the thermal conductivity in this material with non- d -wave order parameter. The finite and universal κ/T at $T \rightarrow 0$ K in the zero impurity limit provides a key support for the SC gap with line nodes. Negligibly small phonon contribution to the total κ allowed us to characterize the T dependence of the electronic κ , which was not possible up to now in the high- T_c cuprates [8]. We found a reasonable agreement with the theoretical predictions by Graf *et al.* [4] in the unitarity scattering limit.

Single crystals of Sr_2RuO_4 were grown in an infrared image furnace by the floating zone method [33]. We used samples of 5 different batches with $T_c = 1.44$ K, 1.32 K, 1.27 K, 1.09 K and 0.71 K, denoted as # 1 - 5, respectively. The T_c and the transition width δT_c were defined at the peak temperature and the full width at half maximum of the imaginary part of the AC susceptibility, respectively (Table I). The difference in T_c comes from the variation of the density of non-magnetic impurities, such as Al or Si [18], and/or crystalline defects [19]. The sys-

tematic studies on the electrical resistivity ρ and resistive T_c revealed that the suppression of T_c is well described by the Abrikosov-Gorkov type equation [34],

$$\ln\left(\frac{T_{c0}}{T_c}\right) = \Psi\left(\frac{1}{2} + \frac{\hbar\Gamma}{2\pi k_B T_c}\right) - \Psi\left(\frac{1}{2}\right), \quad (1)$$

where $\Psi(x)$ is the digamma function, $T_{c0} = 1.5$ K is maximum T_c for the disorder free material and Γ is the non-magnetic impurity scattering rate in the normal state. The values of Γ for the samples under study, deduced from Eq. (1), are listed in Table I.

The thermal conductivity was measured along the in-plane [100] direction between 100 mK and 2 K by the one-heater and two-thermometer method described elsewhere [24]. Typical dimensions of the samples were 2.0 mm \times 0.5 mm \times 0.08 mm, with the longest direction corresponding to the [100] crystal axis and the shortest to the [001] axis.

Fig. 1 shows the T dependence of κ/T of samples # 1 to 5, with the T_c indicated by arrows. All the samples show almost flat T dependence of κ/T in the normal state, as expected for the impurity scattering regime, except for sample # 1 showing slight decrease with T indicative of small electron-electron scattering. The normal state conductivity increases with T_c , i.e. with decreasing Γ . Below T_c , κ/T decreases monotonically and slowly. In sample # 1, κ/T at 0.3 K ($0.2 T_c$) is ~ 5 W/K²m ($0.2 \kappa(T_c)/T_c$), which should be practically zero for the fully gapped superconductor [35]. Although the normal state κ/T decreases 4 times with increasing Γ , in the SC state all κ/T curves converge to a finite value at around 2 ± 0.5 W/K²m at very low T .

The ρ and κ/T measurements made on the same contacts in zero H above T_c revealed that the Wiedemann-Franz ratio $\rho\kappa/T$ at T_c is satisfied within accuracy of our determination ($\pm 10\%$ [36]) for all sample qualities under study, showing a dominance of the electronic transport in the normal state at temperatures as high as T_c . In the SC state, the phonon conductivity κ^g can be estimated in the boundary scattering limit, providing an upper bound for κ^g , by using a simple kinetic formula $\kappa_B^g = \frac{1}{3}C_{ph}\bar{v}l$, where C_{ph} is the phonon specific heat per unit volume proportional to T^3 , \bar{v} is the averaged sound velocity and l is the phonon mean free path, which is of the order of the sample dimension. Since both \bar{v} and l are temperature independent, it can be expressed as $\kappa_B^g/T = \lambda_2^g T^2$ with a constant coefficient λ_2^g . Taking the phonon specific heat coefficient of 0.197 mJ/K⁴mole [21] and $\bar{v}=1400$ m/s [37,38], we obtain $\lambda_2^g = 0.2$ W/K⁴m, which is a small fraction of the total κ in all samples.

On T increase, κ/T vs. T deviates from the universal value, shows an upward curvature up to 0.3 K and then turns into a linear increase at higher T . The κ/T vs. T^2 dependence (Fig. 1 inset) shows that super-linear portion of the κ/T vs. T curve is well reproduced by a polynomial expression $\kappa/T = \lambda_0^e + \lambda_2^e T^2$ (we show only the data for samples # 1 and # 3 for clarity). We should notice here that, despite having the same T^2 dependence

as expected for κ^g/T , the observed κ/T is completely electronic in origin, which is of special interest in this study. Actually measured λ_2^g decreases with decreasing T_c from 44 W/K⁴m for sample # 1 to 8 W/K⁴m for sample # 5, but still is 40 times larger than λ_2^g of 0.2 W/K⁴m. The finite residual coefficient λ_0^e indicates existence of QP at $T=0$ and, consequently, the non- s -wave pairing. In discussion of the residual term which follows below, we use a conventional notation, $\lambda_0^e \equiv \kappa_{00}/T$.

In Fig. 2 we plot (filled squares) κ_{00}/T as a function of the impurity scattering rate divided by the impurity-free T_c , $\hbar\Gamma/k_B T_{c0}$. The vertical error bars reflect the scatter of observed κ/T and the horizontal error comes from the transition width δT_c . For the reference, we show with filled triangles κ/T by Suderow *et al.* [32] extrapolated at $T \rightarrow 0$ K. It can be clearly seen that *the residual thermal conductivity κ_{00}/T remains unchanged for $\hbar\Gamma/k_B T_{c0} < 0.2$* . Moreover, κ_{00}/T values converge to a finite value of 1.7 ± 0.15 W/K²m in the $\hbar\Gamma/k_B T_{c0} \rightarrow 0$ limit, giving strong evidence for the *nodal* gap structure. The finite κ_{00}/T should appear even in the superconductors with fully gapped non- s -wave state (e.g., isotropic 2D p -wave state [20]) when a small number of impurities exist, but it should vanish in the $\Gamma \rightarrow 0$ limit with breaking of the universality [39].

Theoretically, the universal residual term κ_{00}/T for the nodal superconductor is explicitly expressed as

$$\frac{\kappa_{00}}{T} = \frac{\pi^2 k_B^2}{3} N_F v_F^2 \frac{a\hbar}{2\mu\Delta_0}, \quad (2)$$

where N_F and v_F are the density of states in the normal state and the Fermi velocity, respectively. Δ_0 is the maximum amplitude of the gap, μ is the slope of the gap at the node defined as $\mu \equiv \frac{1}{\Delta_0} \frac{\partial \Delta(\phi)}{\partial \phi}$, where ϕ is the in-plane angle of the wave vector on the circular Fermi surface, and a is the coefficient of the order of unity which depends on the gap symmetry [4]. Thus from the residual value κ_{00}/T we can obtain the quantity related to the gap structure, $\mu\Delta_0/a$.

By assuming that the SC gaps have the same magnitude on the three Fermi surfaces of Sr₂RuO₄, and taking experimentally measured Fermi velocities and effective masses for respective bands [16], we can reproduce the observed residual value of $\kappa_{00}/T = 1.7$ W/K²m with $\mu\Delta_0/a = 0.39$ meV. This value is close to typical values for several nodal gap structures. For the 2D f -wave gap with four vertical line nodes $\Delta = \Delta_0 \cos(2\phi)$ [30], a is $4/\pi$ [4], $\mu = 2$, $\Delta_0 = 2.14 k_B T_c$ in the weak coupling limit and we obtain $\mu\Delta_0/a = 0.43$ meV. For the state with horizontal line node, indicated in recent directional experiments [25–27], the universal thermal conductivity was not considered theoretically. However, we can estimate the residual value of κ_{00}/T by assuming that in the limit of the small Fermi velocity along the [001] axis $v_{F\perp} \rightarrow 0$, the 3D polar state $\Delta = \Delta_0 \cos(\theta)$ (θ is polar angle measured from the [001] axis) is a good representation of a Q2D horizontal nodal state. By using the parameters

for the polar state [4] $a=1$, $\mu = 1$ and $\Delta_0 = 2.46k_B T_c$, we obtain $\mu\Delta_0/a = 0.32$ meV, which is also close to the experimentally observed value. Thus we conclude that the observed residual term comes from the contribution around the gap nodes.

Recently Zhitomirsky and Rice [31] proposed the orbital dependent gap structure, which is a modified extension of the original theory by Agterberg *et al.* [28]. They assumed the line nodes only on two of three Fermi surfaces, α and β . If we consider that only these two sheets make contribution to $\kappa_{00}/T = 1.7$ W/K²m, we obtain $\mu\Delta_0/a = 0.27$ meV. They also assumed a very small sub-Kelvin amplitude of the gap Δ_0 on these sheets; thus to explain the observed value of κ_{00}/T we need a relatively steep slope μ at the nodes.

Since the observed κ/T is almost purely electronic, we can compare the experimental results with available theories for the electronic thermal conductivity. Two theories predict a T^2 term in the electronic κ/T . The model by Zhitomirsky and Walker [40] and by Graf and Balatsky [41] (ZWGB) is valid in the range of $\gamma_{\text{imp}} < k_B T$ due to the QP density coming from the nodal SC gap in case of unitary scattering. The model by Graf *et al.* [4] considers finite T correction to the residual conductivity κ_{00}/T in the range of $k_B T < \gamma_{\text{imp}}$, where the coefficient of the T^2 term of κ/T strongly depends on the impurity scattering phase shift [4]. As can be seen from Inset in Fig. 1, the T^2 term is observed from the lowest T , directly above universality limit. This implies that the condition $\gamma_{\text{imp}} < k_B T$ required for observation of the T^2 term in ZWGB model is not met. Therefore, we stick in interpretation to the Graf model [4]. Here the T^2 term is observed as long as we satisfy the condition for $k_B T < \gamma_{\text{imp}}$, which corresponds to $T < 0.3$ K in our experiment. This relatively large impurity band means that the scattering is strong. Indeed, in the unitarity limit (the phase shift $\delta = \pi/2$) the impurity bandwidth increases rapidly with the impurity scattering rate as $\gamma_{\text{imp}} \sim \sqrt{\Delta_0 \hbar \Gamma}$ [2], which corresponds to about 0.5 K for sample # 1, while in the Born limit ($\delta = 0$) the impurity bandwidth becomes exponentially small, $\gamma_{\text{imp}} \sim \Delta_0 \exp(-\frac{\Delta_0}{\hbar T})$ [2]. Experimentally, the impurity bandwidth can be estimated from the parameters of the fitting formula, $\kappa/T = \lambda_0^e + \lambda_2^e T^2$ introduced above. In the unitarity scattering limit, the deviation of κ/T from the universal value at finite T is described as [4]

$$\frac{\kappa}{T} = \frac{\kappa_{00}}{T} \left(1 + \frac{7\pi^2}{60} \left(\frac{k_B T}{\gamma_{\text{imp}}} \right)^2 \right). \quad (3)$$

The γ_{imp} deduced by comparing Eq. (3) with the coefficients λ_0^e and λ_2^e for each sample is plotted in Fig. 2 with an open circle. The uncertainty due to the possible small phonon contribution is at most 10 %. The least square fit with the \sqrt{T} function $\gamma_{\text{imp}} = 0.7\sqrt{\hbar \Gamma/k_B T_{c0}}$ is drawn with a dotted line. The rapid increase and \sqrt{T} dependence of γ_{imp} support the strong impurity scattering and verify the deduction of γ_{imp} from Eq. (3), while

the absolute value is about two times smaller than the theoretical value of $\gamma_{\text{imp}} \sim \sqrt{\Delta_0 \hbar \Gamma}$. A slight deviation of δ from $\pi/2$ makes the impurity bandwidth exponentially small at small Γ [4] and thus the non-negligible γ_{imp} of sample # 1 ($\hbar \Gamma/k_B T_{c0} = 0.051$) implies the large scattering phase shift $\delta = \pi/2(\pm 10\%)$.

From our data we can derive the explicit condition of the low T and small γ_{imp} "universality limit" which is theoretically described as $k_B T \leq \gamma_{\text{imp}} \ll \Delta_0$. In Fig. 2 we can see that above $\hbar \Gamma/k_B T_{c0} = 0.2$ the universality starts to break and this crossover point corresponds to $\gamma_{\text{imp}}/k_B \sim 0.3$ K $\sim 0.1\Delta_0/k_B$. Thus $k_B T \leq \gamma_{\text{imp}} \leq 0.1\Delta_0$ can be considered as an experimentally deduced criterion for the observation of universal conductivity.

The deviation of κ_{00}/T from the universal value with increasing $\hbar \Gamma/k_B T_{c0}$ is small, even for the largest $\hbar \Gamma/k_B T_{c0}$, giving at most 50 % increase for sample # 5 ($\hbar \Gamma/k_B T_{c0} = 0.6$). According to the calculation by Sun and Maki [3] in the unitarity scattering limit, κ_{00}/T increases rapidly with Γ above the universality limit and it reaches almost twice of the universal value at $\hbar \Gamma/k_B T_{c0} = 0.6$. Similar slow increase of κ_{00}/T was also pointed out in YBa₂Cu₃O_{6.9} [6]. While the slight deviation of the scattering phase shift from $\pi/2$ might be able to explain it [6], the calculated steep increase of κ_{00}/T [3] is inappropriate for the description of Sr₂RuO₄. The critical resistivity, above which T_c disappears, is about $1 \mu\Omega\text{cm}$ [18,19] and the corresponding critical κ/T is obtained by the Wiedemann-Franz law as 2.4 W/K²m, which is almost equal to our largest κ_{00}/T . Since there is no clear reason for κ/T at $T=0$ K to become higher in the SC state than in the normal state, the increase of κ_{00}/T with Γ expected in Ref. [3] should be limited. This point should be clarified in the further studies.

In conclusion, we experimentally observed universal thermal conductivity in Sr₂RuO₄, indicating that this phenomenon is not restricted to d -wave gap symmetries. The universality gives clear evidence for the nodal gap. From the analysis of the low temperature thermal conductivity within the framework of the theory by Graf *et al.* [4], we extract the impurity bandwidth and the scattering phase shift close to $\pi/2$. We experimentally derive a criterion for the deviation from the universal limit.

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TABLE I. The properties of 5 single crystals of Sr_2RuO_4 . T_c and δT_c were determined from the AC susceptibility measurements. The impurity scattering rate normalized by the maximum T_c , $\hbar\Gamma/k_B T_{c0}$ is deduced from Eq. (1).

	# 1	# 2	# 3	# 4	# 5
T_c (K)	1.44	1.32	1.27	1.09	0.71
δT_c (K)	0.02	0.03	0.03	0.05	0.15
$\hbar\Gamma/k_B T_{c0}$	0.051	0.15	0.20	0.35	0.60

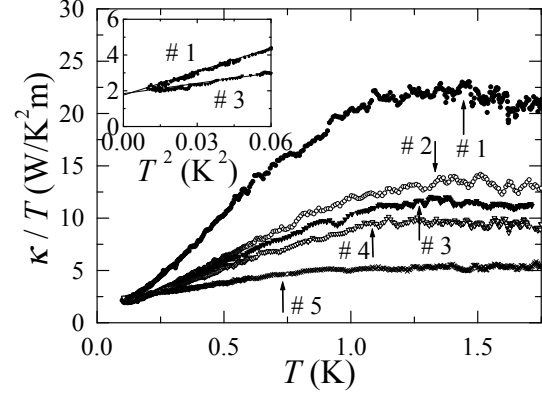


FIG. 1. The temperature dependence of the thermal conductivity κ/T along the [100] direction for 5 samples of different quality. The arrows denote the transition temperatures T_c . Inset shows the κ/T vs. T^2 plot for samples # 1 and # 3.

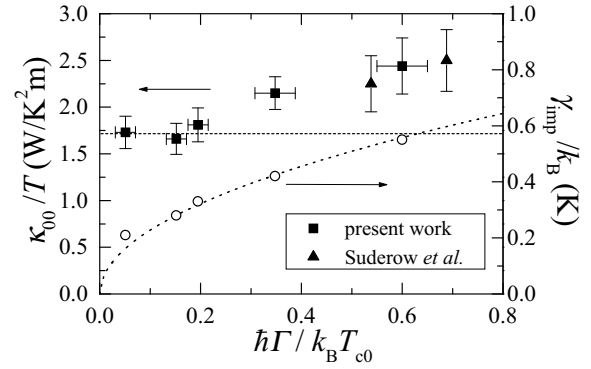


FIG. 2. The residual thermal conductivity κ_{00}/T and the impurity bandwidth γ_{imp} as a function of scattering rate $\hbar\Gamma/k_B T_{c0}$. Filled squares show κ_{00}/T of this study, filled triangles are by Suderow *et al.* [32]. The open circles represent γ_{imp} and the dotted line is a fit with the \sqrt{T} function.